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This report describes the progress on the development of a new laser-based combustion diagnostic for the quantitative measurement of both major and minor species in high-pressure flames. The technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond pump/probe spectroscopy. A method is presented for vastly improving the output of the synchronously mode-locked dye laser systems. A pump/probe absorption model is used to estimate the detection limit. A new differential detector is described. A modification is made to the basic instrument to achieve shot-noise-limited detection.

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ASYNCHRONOUS OPTICAL SAMPLING FOR LASER-BASED COMBUSTION DIAGNOSTICS IN HIGH-PRESSURE FLAMES

Progress Report
Air Force Office of Scientific Research
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Abstract

This report describes the progress on the development of a new laser-based combustion diagnostic for the quantitative measurement of both major and minor species in high-pressure flames. The technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond pump/probe spectroscopy. A method is presented for vastly improving the output of the synchronously mode-locked dye laser systems. A pump/probe absorption model is used to estimate the detection limit. A new differential detector is described. A modification is made to the basic instrument to achieve shot-noise-limited detection.

1. RESEARCH OBJECTIVES

The overall goal of this research is to develop and test a new combustion diagnostic for the quantitative measurement of both major and minor species in high-pressure flames. The proposed technique, Asynchronous Optical Sampling (ASOPS), is a state-of-the-art improvement in picosecond laser spectroscopy which should yield a better signal-to-noise ratio (SNR) than laser fluorescence measurements in rapidly quenched combustion environments. Furthermore, ASOPS will allow determination of both quenching rates and state-to-state relaxation rates which are necessary for quantitative applications of both laser-induced and laser-saturated fluorescence at high pressure. The ASOPS technique produces a coherent signal-carrying beam and thus requires no more optical access to practical combustion devices than LDV measurements.

2. RESEARCH STATUS

2.1 Progress to Date

In past experiments, visible ASOPS studies of atomic sodium in an atmospheric-pressure flame proved to be quite successful. In order to evaluate the difficulty of using UV beams, the next subject of study was atomic indium. Although a signal was obtained, the signal-to-noise ratio (SNR) was less than expected. Experimental detection of the hydroxyl radical using ASOPS was not successful, despite our early estimates of SNR, and despite several attempts at reducing the noise emitted from the laser systems. Research during the past year has consisted, in part, of a systematic procedure to explain exactly why the above scenario was observed.

The first goal of the past year's research was to examine more closely the nature and causes of noise emitted from the dye lasers. This has been completed, and it is now known why the UV experimental results were less favorable than expected. More importantly, this study has led to a simple, inexpensive method for reducing noise on the dye laser output by orders of magnitude. The second goal was to develop a quantitative model that relates the detected modulation of the probe beam to an absolute concentration in the flame. This has also been completed, and will be summarized below. In past research, differential detection circuits have been shown in our laboratory to reduce noise by ~ 20 dB. The drawback to these devices was that long-term drift in laser power reduced the noise subtraction effectiveness, requiring constant readjustment of the power of the input beams. A new detector that automatically compensates for long-term drift has been constructed and tested. The results are presented below.

2.1.1 Improvement of Laser Performance

For shot-noise-limited detection, it is has been reported that a minimum detectable probe modulation depth of $\sim 10^{-8}$ can be detected.[1] From our own studies of dye-laser noise, and from similar results obtained by other investigators, we know that the shot noise limit is approached only at frequencies beyond several megahertz.[1,2,3] Previous ASOPS studies were carried out at a beat frequency near 10 kHz, where the noise level is approximately 10^3-10^4 larger than the shot-noise limit. In estimating the SNR of hydroxyl experiments, we previously assumed that the noise on the UV beams was identical to that on the fundamental dye-laser output. We now know that the noise on the frequency doubled beam is significantly larger, and extends out to roughly twice the bandwidth of the visible dye-laser beam.[4] It is thus not surprising that UV ASOPS detection of indium resulted in a much lower SNR than similar detection of sodium, despite the similarity in absorption cross sections of these atoms.

Previous attempts to reduce dye-laser noise have been hampered by the inability to attribute the noise to a particular cause. Recently, investigators have found that much of the noise is caused by the effects of spontaneous emission on the laser pulse as it propagates through the jet of the dye laser.[5,6,7,8] This has led to a simple, inexpensive feedback method for significantly improving the quality of the laser output.[7,8] The coherent photon feedback technique has been added to our dye lasers, as shown in Fig. 1. The principle is to feedback a secondary laser pulse so that it arrives at the dye jet slightly before the main pulse. The secondary pulse overwhelms any effects that may arise from spontaneous emission. An example of the effects of this technique is shown in Fig. 2. The lowest curve is the unstabilized autocorrelation trace, consisting of two distinct regimes; the lower envelope has a width of 23 psec (FWHM), over which a sharp spike rests. The spike is caused by random pulse-to-pulse fluctuations, indicating that mode-locking is far from optimum. The other two autocorrelation traces were obtained with feedback. The top trace resulted when a feedback power ratio of approximately 0.1 was utilized. As shown, the pulsewidth is quite broad, with a width of 41 psec. The middle trace resulted when a ND filter (88%) was placed in the feedback path. The resulting pulsewidth is 26 psec.

Two other important benefits are realized by using the coherent photon feedback stabilization technique.[5,8] First, transform limited pulses result, and this will allow absolute concentrations to be obtained from ASOPS data, since knowledge of the laser bandwidth is required. Second, the laser pulsewidth can be increased by increasing the power of the feedback pulses. Thus the bandwidth will undergo a corresponding decrease, thereby increasing the ASOPS signal.

The dye laser also contributes to noise from dye jet imperfections. New sapphire dye jets have shown a dramatic decrease in the noise bandwidth compared to specially constructed stainless-steel jets.[9] A sapphire jet nozzle has been added to each of our dye lasers. In addition to increasing the stability of the dye-laser output, the dye laser has been found to be easier to operate with this improvement.

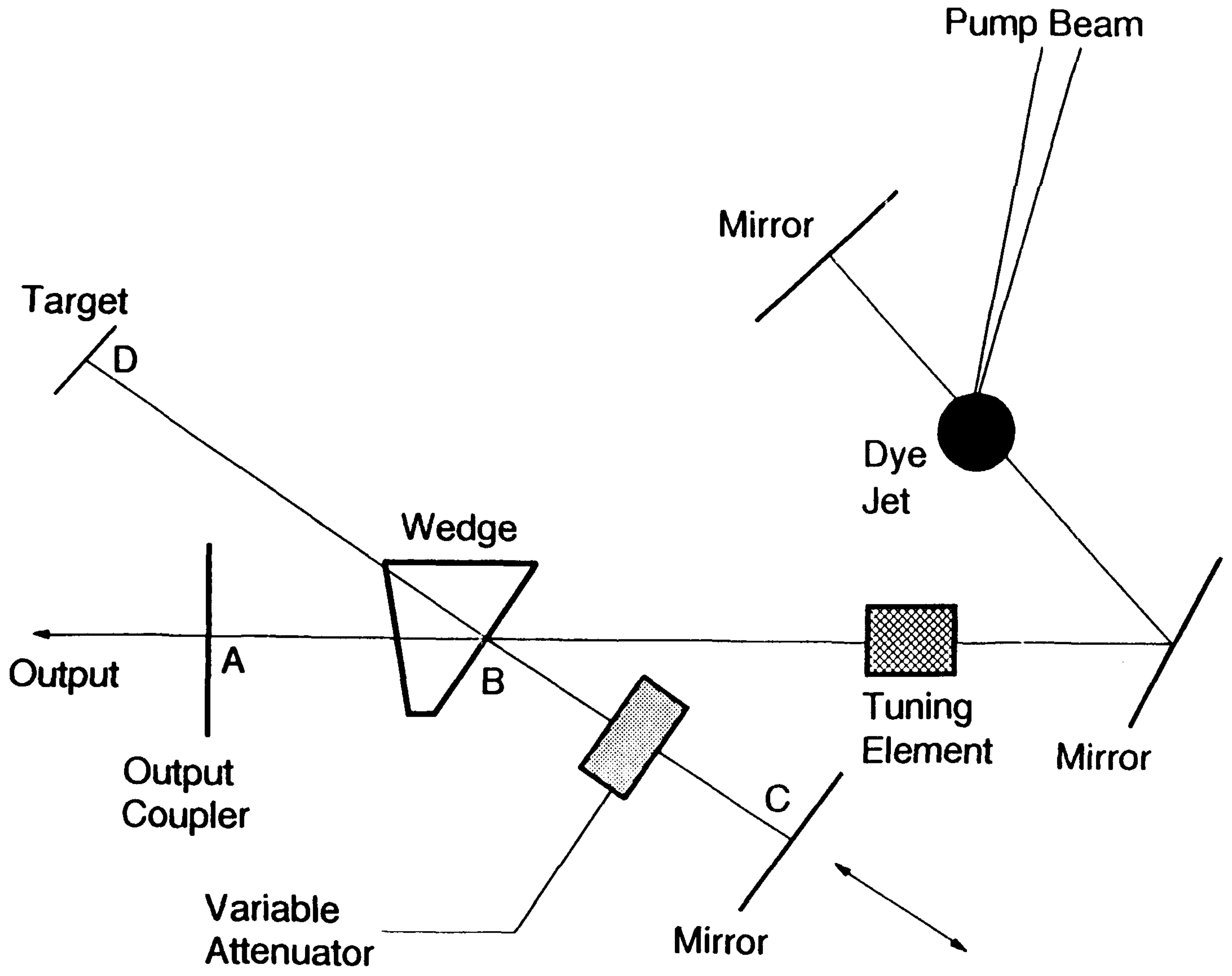

Figure 1. Synchronously mode-locked dye laser with the cavity modified to incorporate coherent photon seeding stabilization.[8]

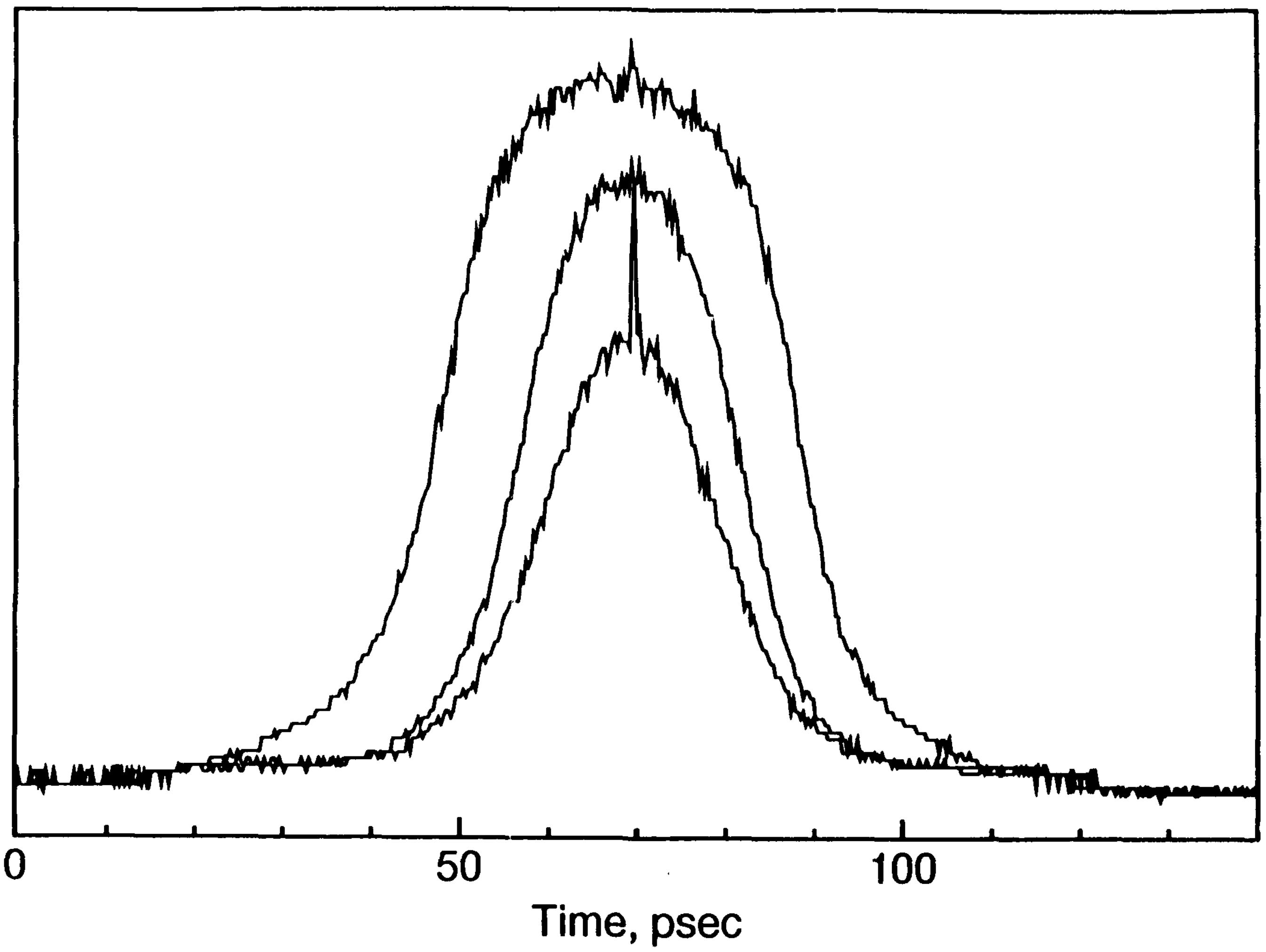

Figure 2. Three autocorrelation traces that demonstrate the effects of coherent photon seeding stabilization. All curves were taken with the dye laser cavity lengthened from the point at which the minimum SHG autocorrelation pulsewidth is produced. The lower curve was obtained without stabilization. The top trace was obtained with a feedback power ratio of approximately 0.1. The middle trace was obtained after a ND filter (88%) was placed in the feedback path.

2.1.2 A Pump/Probe Absorption Model

Using rate equation theory, we have developed a model to predict the modulation depth on the probe laser at the beat frequency as the result of stimulated absorption and stimulated emission. The peak modulation depth was found to take the form

$$\alpha_{\mathrm{mod}} = \left(\frac{g_3}{g_1}\right) \frac{c^4 A_{21} A_{32} P_{\mathrm{AVE}}^{\mathrm{pump}} N_T L}{4\pi^5 D^2 h \nu_{12}^3 \nu_{23}^2 f^L (\Delta\nu_{1/2}^L)^2}, \qquad (1)$$

where c is the speed of light (cm/s), h is Planck's constant (J·s), P_{AVE}^L is the average pump laser power (W), D is the focal diameter of the pump beam (cm), f^L is the pump laser repetition rate (s^{-1}), A_{21} and A_{32} are the Einstein rate coefficients for spontaneous emission of the transitions in resonance with the pump laser and the probe laser, respectively (s^{-1}), and $\Delta\nu_{1/2}^L$ is the spectral width (FWHM) of the laser (s^{-1}), which is assumed to be identical for each beam. The degeneracy of the state from which the pump removes molecules is given by g_1, while g_3 denotes the degeneracy of the state to which the probe places molecules.

Assuming a minimum detectable modulation depth of 10^{-8} can be achieved, Eqn. (1) can be used to estimate detection limits for the ASOPS technique. For atomic sodium, with a focal diameter of 50 μm, and pump and probe beams crossed at an angle of 5°, a detection limit of 3.3×10^6 cm^{-3} is estimated. For the $Q_1(9)$ line of OH in a flame at 2000 K, a detection limit of 8.3×10^{12} cm^{-3} is predicted. It is thus easy to see why previous attempts at detection of OH failed, while detection of atomic sodium was successful. Not only is the dye laser noise substantially worse when the laser is frequency doubled, the pump/probe absorption interaction is much weaker. Nevertheless, these results show that if the shot-noise limit can be attained, OH detection is possible.

2.1.3 Improvement of Detection Electronics

The automatic subtraction circuit shown in Fig. 3 has been constructed for differential detection of the probe beam with the ASOPS instrument.[10] This circuit removes probe beam fluctuations from the signal by subtracting the signal beam intensity, detected leaving the test section, from a sample beam split off prior to the test section. The circuit is an improvement over previous differential detection circuits since it eliminates the need to match signal and sample beam powers. The circuit uses a closed loop integrating servo amplifier to automatically adjust the current splitting between the signal and sample photocurrents. Figure 4 shows the noise spectrum of a mode locked synchronously pumped dye laser measured using the automatic subtraction circuit and a 20-kΩ transimpedance amplifier. The automatic subtraction circuit reduces noise by as much as 15 dB at low frequencies compared to the transimpedance amplifier.

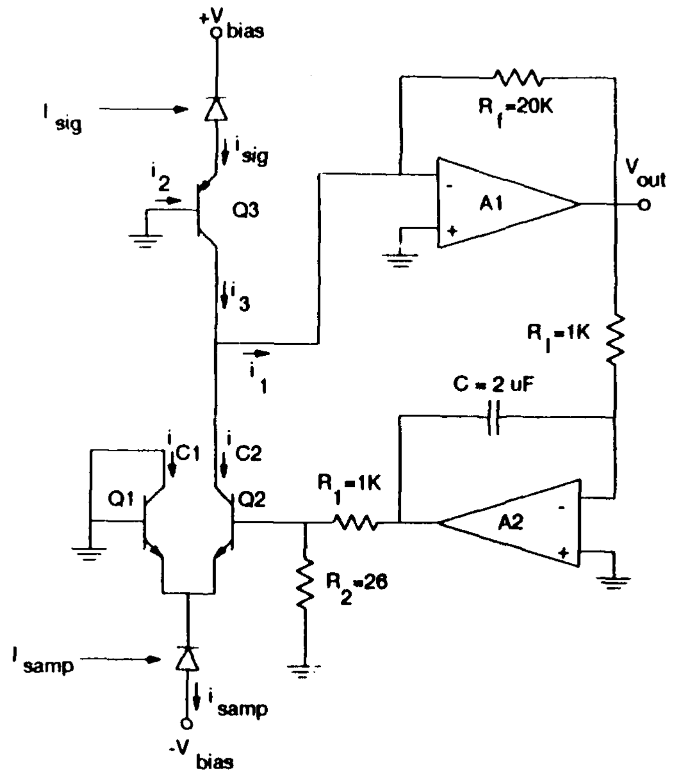

Figure 3. Automatic subtraction circuit diagram.[10] The following symbols are used in the circuit diagram: A1 and A2 denote OP27GP operational amplifiers; Q1 and Q2 denote MAT-04FP matched transitors; and Q3 denotes a 2N3906 transitor. The photodiodes are Hamamatsu 1226S-18BQ.

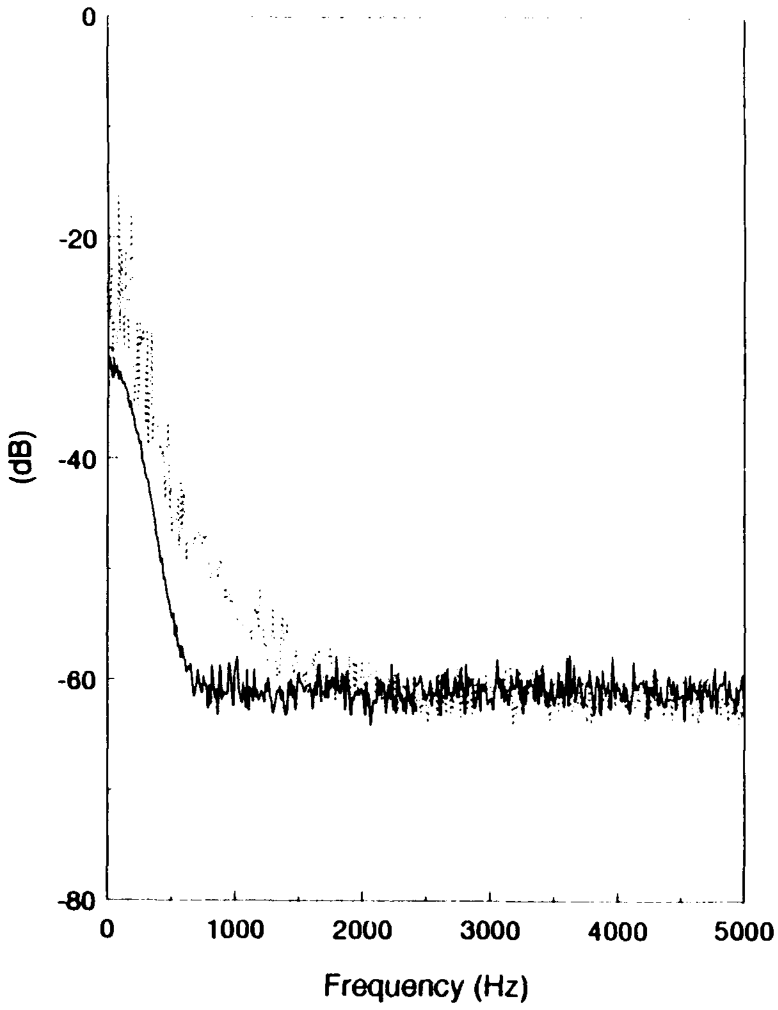

Figure 4. Noise spectra that demonstrate the automatic subtraction circuit. The lower trace was obtained using the subtraction circuit, while the upper trace was obtained using a photodiode in conjunction with a 20-kΩ transimpedance amplifier. The two curves are matched by aligning the peaks of a 10-kHz amplitude modulation measured for both curves (outside plotted range).

2.2 Current Research

Knowing that OH detection will be possible if the shot-noise limit can be achieved, current research is directed toward modifying the basic ASOPS instrument. An electrooptic modulator (Quantum Technology Model 3010) has been added to the pump laser. This makes it possible to shift the ASOPS harmonics beyond the noise corner frequency, as shown in Fig. 5, where the ASOPS signal appears as sidebands about the electrooptic modulation frequency. The modulated ASOPS (MASOPS) instrument diagram is shown in Fig. 6. The two independently mode-locked laser systems of the conventional ASOPS instrument remain intact. To maintain a constant phase walk-out between the pump and probe pulse trains, the frequency synthesizers of each laser are operated in a master-slave configuration, referenced to the same 10-MHz oscillator. To ensure a constant number of pump and probe beam pulses per modulator cycle, the frequency synthesizer for the modulator is also referenced to the 10-MHz oscillator. After the pump and probe lasers are crossed in the flame, the probe is again directed to a suitable detection system, and the output of this is filtered and amplified.

The MASOPS signal must then be demodulated, so that the temporal information that is contained in the sidebands can be analyzed. This is accomplished using the frequency downconverter detection system of Fig. 7.[11] The output of a second 10-MHz oscillator is split into two portions. One portion is mixed with half of the modulator frequency-synthesizer output, which can be set at 9.95 MHz. The other half of the 10-MHz synthesizer output is mixed with the filtered and AC-amplified signal. The resulting 50-kHz signal is detected with a low-frequency lock-in amplifier (Stanford Research Systems SR510), triggered by the down-shifted modulator reference signal. The output of the lock-in amplifier is identical to the ASOPS signal of our previous instrument, and is thus directed to a digitizing oscilloscope, which is triggered at the beat frequency.

2.3 Future Work

When the frequency downconverter detection system has been constructed, the MASOPS instrument will be initially tested by again detecting atomic sodium. This will be aided by the familiarity we have with the detection of this atom. Using the pump/probe absorption model, absolute concentrations will be obtained, and the detection limit will be evaluated. From these results, it will be possible to determine if the shot noise limit has been approached using the MASOPS detection scheme. Upon completion of the visible MASOPS studies, frequency doublers will again be added to the pump and probe beams, and OH will be detected.

3. PUBLICATIONS AND PRESENTATIONS

The following paper has been submitted for publication.

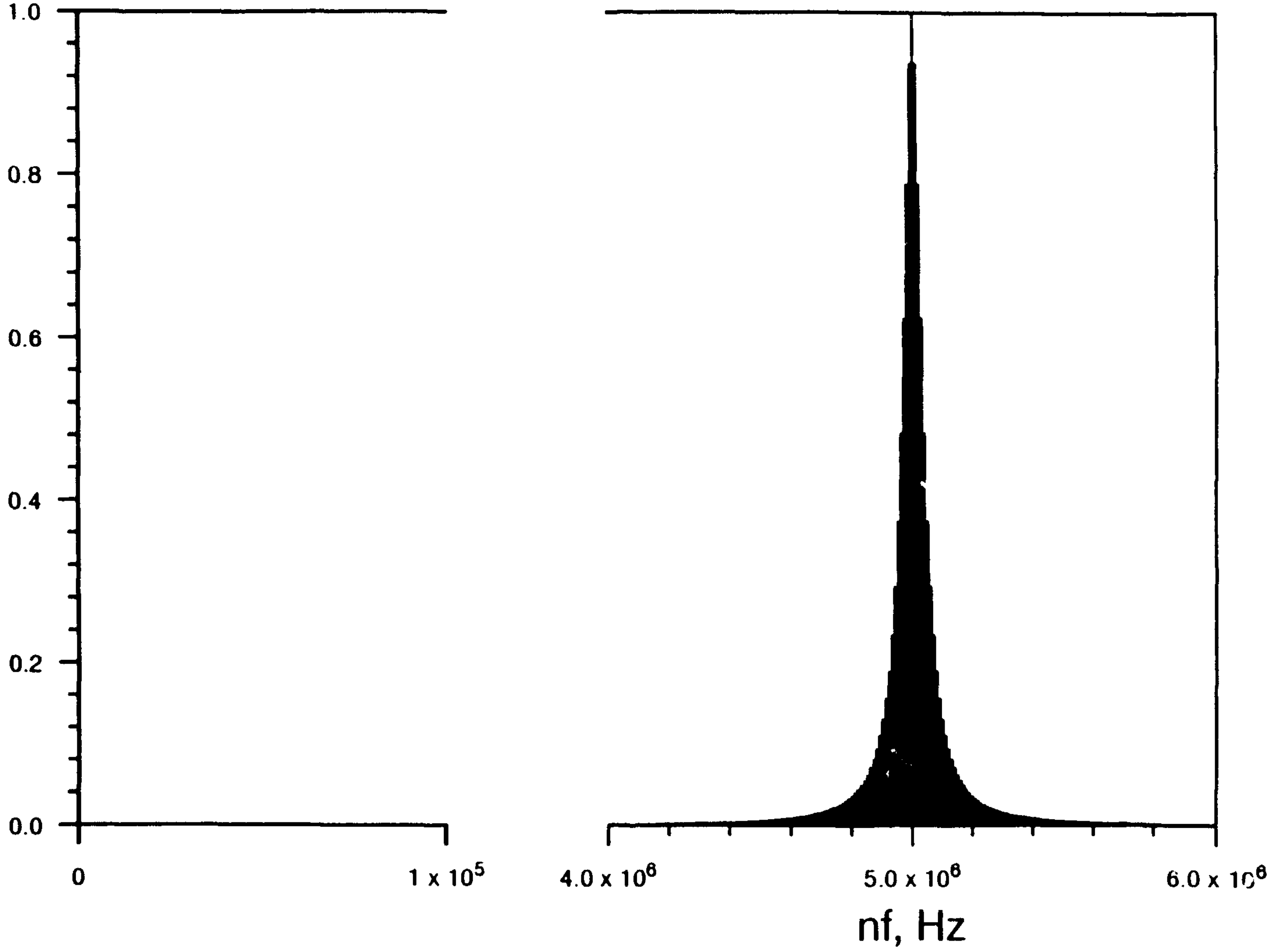

Figure 5. Spectrum that results when the ASOPS signal is obtained with a sinusoidally modulated pump beam. The modulation frequency is 5 MHz in this example.

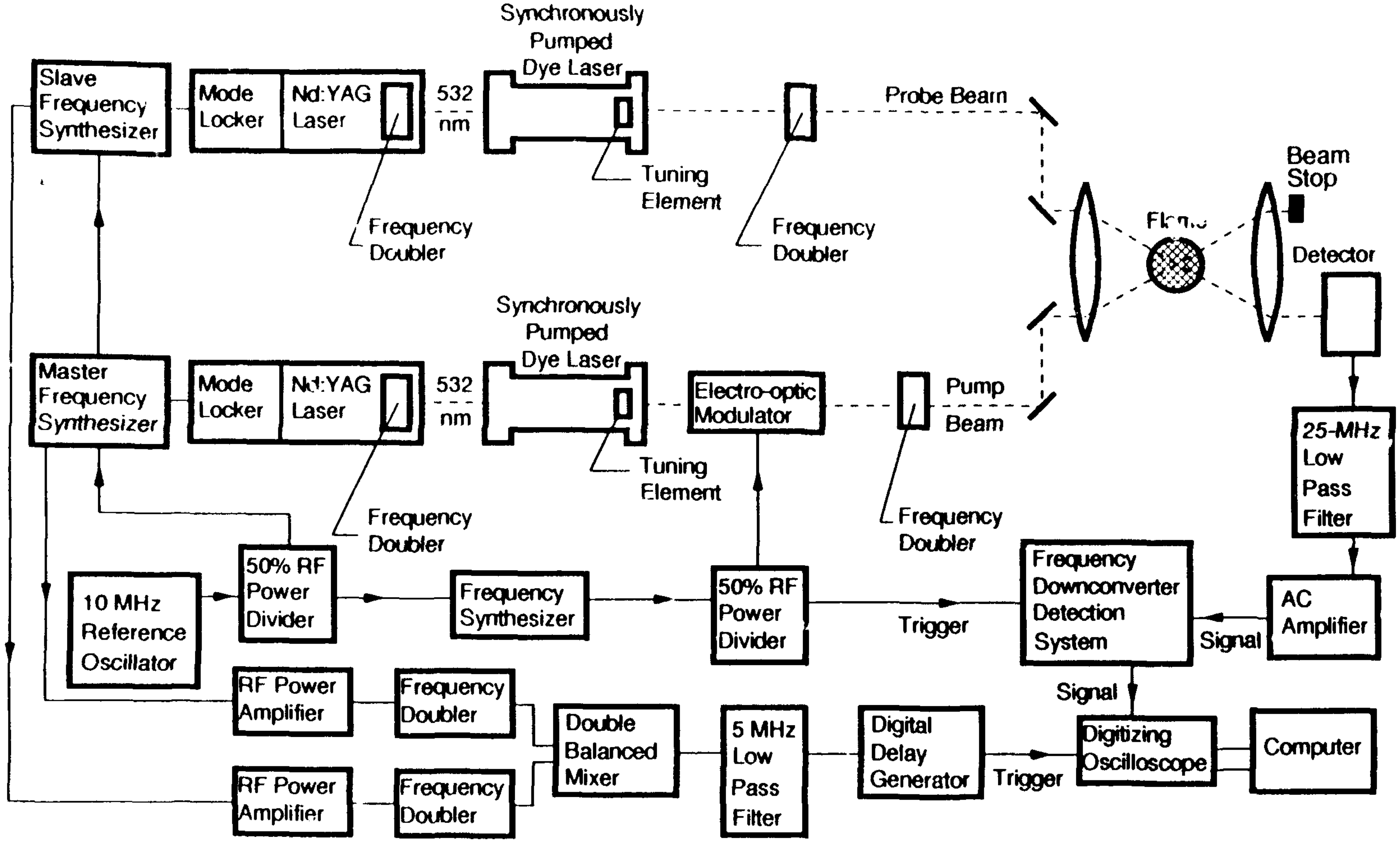

Figure 6. Modulated ASOPS (MASOPS) detection instrument.

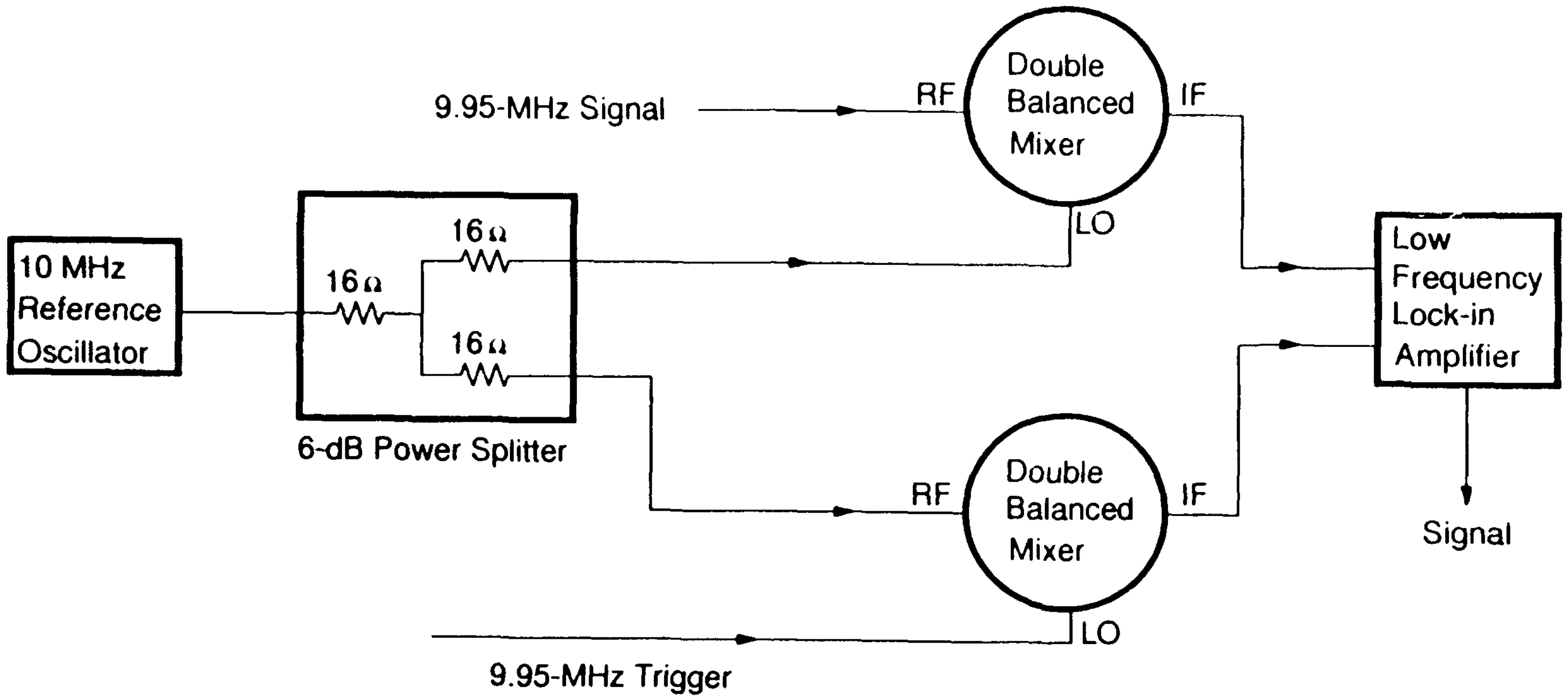

Figure 7. Frequency downconverter detection system.[11]

1. G. J. Fiechtner, G. B. King, N. M. Laurendeau, and F. E. Lytle, "Determination of Relative Number Density and Decay Rate for Atomic Sodium in an Atmospheric Premixed Flame by Asynchronous Optical Sampling", *submitted to Applied Optics.*

4. RESEARCH PERSONNEL

Professors Galen B. King and Normand M. Laurendeau in the School of Mechanical Engineering and Professor Fred E. Lytle in the Department of Chemistry are co-principal investigators for this research. Dr. Ronald Kneisler completed his Ph.D. thesis in September of 1990, and has left the group. Mr. Gregory Fiechtner, a mechanical engineering Ph.D. candidate, joined the group in July, 1986 as a M.S. candidate, completing his M.S. thesis in August of 1989. He continues on the ASOPS project as a Ph.D. candidate. Mr. Brian Thompson joined the group in August of 1990 as an M.S. candidate in mechanical engineering.

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